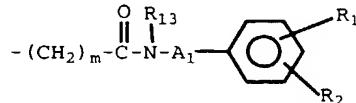


What is claimed is:

1. A diagnostic agent comprising an aminocarboxylate ligand complexed with a paramagnetic metal ion wherein a nitrogen atom within said 5 aminocarboxylate is substituted with a substituted aromatic amide group.
2. The diagnostic agent of claim 1 wherein said substituted aromatic amide group is of the formula

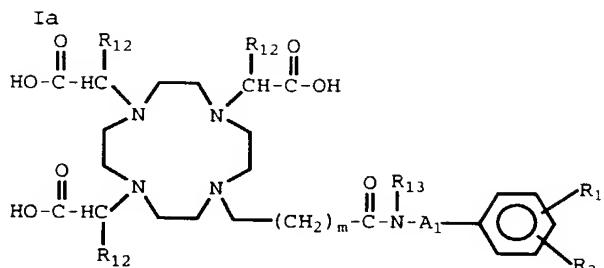
10 I



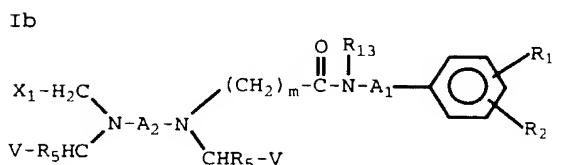
wherein

- A₁ is -(CH₂)_m'- or a single bond;
- (CH₂)_m and (CH₂)_m' may independently be 15 substituted with alkyl or hydroxyalkyl;
- R₁ and R₂ are each independently hydrogen, 20 alkyl, -NO₂, -NH₂, -NHCNHR₁₂, NCS, -C(=O)-NR₃R₄, NR₃COR₉ where R₉ is alkyl or hydroxyalkyl, with the proviso that at least one of R₁ and R₂ must be other than hydrogen;
- R₃ and R₄ are independently hydrogen, alkyl, arylalkyl, aryl, alkoxy and hydroxyalkyl;
- R₁₂ is hydrogen, alkyl or hydroxyalkyl;
- R₁₃ is hydrogen, alkyl, arylalkyl, aryl, 25 alkoxy or hydroxyalkyl;
- m and m' are independently 1 to 5; and multimeric forms thereof.

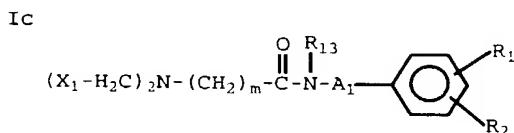
3. A diagnostic agent of claim 2 wherein said ligand is of the formula



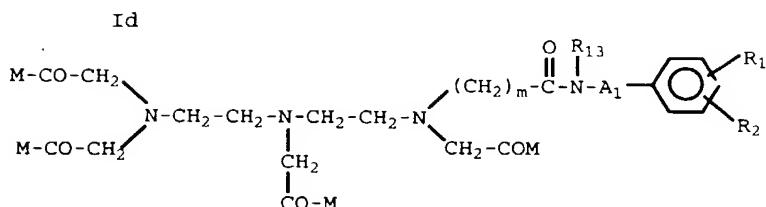
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wherein m, R13, A1, R1, R2, and R12 are as defined in claim 2 and wherein

X_1 is $-COOY_1$, PO_3HY_1 or $-CONHOY_1$;

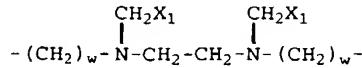
Y_1 is a hydrogen atom, a metal ion equivalent and/or a physiologically biocompatible cation of an inorganic or organic base or amino acid;

V is X₁ or is -

wherein X_1 is as defined above, B is a protein or

20 V is X_1 or is $-CH_2OH$, $-CONH(CH_2)_rX_1$ or $-COB$,
wherein X_1 is as defined above, B is a protein or
lipid residue, r is an integer from 1 to 12, or if
 R_5 , R_6 and R_7 are each hydrogen; then both V's
together form the group

25



where X_1 is as above, w is 1, 2 or 3, provided that at least two of the constituents Y_1 represent metal

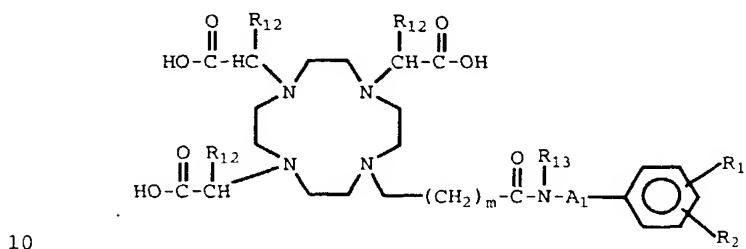
- 30 ion equivalents of an element with an atomic number of 21 to 29, 42, 44 or 57 to 83; from 1 to 4, advantageously 2 or 3, and preferably 2 M's are -OH and the balance independently are -OR₁₀, -NH₂,

-NHR₁₀ and/or NR₁₀R_{10'} wherein R₁₀ and R_{10'} are selected from an organic alkyl radical of up to 18 carbon atoms which may be substituted.

4. The diagnostic agent of claim 1 wherein
5 said paramagnetic metal ion is gadolinium.

5. A compound of formula Ia, Ib, Ic or Id as defined in claim 3, including multimers thereof.

6. A compound of the formula



wherein

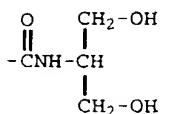
- A₁ is -(CH₂)_m'- or a single bond;
(CH₂)_m and (CH₂)_m' may independently be
15 substituted with alkyl or hydroxyalkyl;
R₁ and R₂ are each independently hydrogen,
alkyl, -NO₂, -NH₂, -NHCNHR₁₂, NCS, -C(=O)-NR₃R₄ and
NR₃COR₉ where R₉ is alkyl or hydroxyalkyl, with the
proviso that at least one of R₁ and R₂ must be other
20 than hydrogen;
R₃ and R₄ are independently hydrogen, alkyl,
arylalkyl, aryl, alkoxy and hydroxyalkyl;
R₁₂ is hydrogen, alkyl or hydroxyalkyl;
R₁₃ is hydrogen, alkyl, arylalkyl, aryl,
25 alkoxy or hydroxyalkyl;
m and m' are independently 1 to 5;
and multimeric forms thereof.

7. A compound of claim 6 wherein R₁ and R₂ are each $\begin{array}{c} \text{O} \\ \parallel \\ -\text{C}-\text{NR}_3\text{R}_4 \end{array}$ wherein each R₃ group is hydroxy-alkyl.

8. A compound of claim 6 wherein R₁ and R₂ are each $\begin{array}{c} \text{O} \\ \parallel \\ -\text{C}-\text{NR}_3\text{R}_4 \end{array}$ wherein each R₃ group is selected from $\begin{array}{c} \text{OH} \\ | \\ -\text{CH}_2-\text{CH}-\text{CH}_2\text{OH} \end{array}$ and $\begin{array}{c} \text{OH} \\ | \\ -\text{CH}(\text{CH}_2\text{OH})_2 \end{array}$, and wherein each R₄ group is hydrogen.

9. A compound of claim 6 wherein R₁ and R₂ are each $\begin{array}{c} \text{O} \quad \text{OH} \\ \parallel \quad | \\ -\text{CNHCH}_2-\text{CH}-\text{CH}_2-\text{OH} \end{array}$.

10. A compound of claim 6 wherein R₁ and R₂ are each



11. A compound of claim 6 having the name
15 10-[2-[[3,5-bis([(2,3-dihydroxypropyl)amino]- carbonyl]phenyl)amino]-2-oxoethyl]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

12. The gadolinium complex of the compound of claim 11.

20 13. A compound of claim 6 having the name
10-[2-[[3,5-bis-[[[2-hydroxy-1-(hydroxymethyl)- ethyl]amino]carbonyl]phenylamino]2-oxoethyl]- 1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

25 14. The gadolinium complex of the compound of claim 13.

15. A compound of claim 6 having the name
10-[2-[methyl[3,5-bis[(2-methylbutyl)amino]-

carbonyl]phenyl]amino]-2-oxoethyl]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

16. The gadolinium complex of the compound of claim 15.

5 17. A compound of claim 6 having the name 10-[2-[[4-[[2,3-dihydroxypropyl]amino]carbonyl]-phenyl]amino]-2-oxoethyl-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

10 18. The gadolinium complex of the compound of claim 17.

19. A compound of claim 6 having the name 10-[N-(4-nitrophenyl)acetamido]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

15 20. The gadolinium complex of the compound of claim 19.

21. A compound of claim 6 having the name 10-[N-(4-aminophenyl)acetamido]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

22. The gadolinium complex of the compound of 20 claim 21.

23. A compound of claim 6 having the name 10-[[N-(4-(N'-isothiocyanato)phenyl)acetamido]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

24. The gadolinium complex of the compound of 25 claim 23.

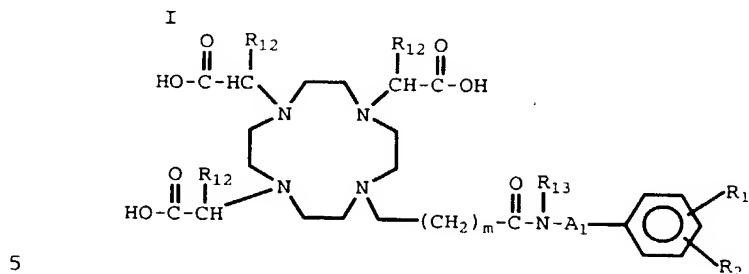
25. A compound of claim 6 having the name 10-[N-[4-(N'-methylthioureido)phenyl]acetamido]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

26. The gadolinium complex of the compound of 30 claim 25.

27. A compound of claim 6 having the name 10-[N-[4-(N',N'-diethylaminothioureido)phenyl]acetamido]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.

28. The gadolinium complex of the compound of claim 27.
29. A compound of claim 6 having the name 10,10'[[{{(1,2-ethanediyl)diimino}bis(thioxomethyl)}-diimino]bis(4,1-phenylene)diimino-bis[1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid].
30. The gadolinium complex of the compound of claim 29.
31. A compound of claim 6 having the name 10,10'-{{(Thioxomethyl)bis(imino)bis(4,1-phenylene)}bis(imino)}bis(2-oxo-2,1-ethanediyl)]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.
32. The gadolinium complex of the compound of claim 31.
33. A compound of claim 6 having the name 10,10',10''-{{{{iminobis(2,1-ethanediyl)triiimino}-tris(thioxomethyl)}-triiimino}tris-(4,1-phenylene)}-triiimino]tris(2-oxo-2,1-ethanediyl)]tris[1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid].
34. The gadolinium complex of the compound of claim 33.
35. A compound of claim 6 having the name 10-[2-[(2-(4-nitrophenyl)ethyl)amino]-2-oxoethyl]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid.
36. The gadolinium complex of the compound of claim 35.
37. A compound of claim 6 having the name 10-[2-[[3,5-bis[(2-hydroxyethyl)amino]-carbonyl]-phenyl]amino]-2-oxoethyl]-1,4,7,10-tetraazacyclododecane-1,4,7-triacetic acid, monosodium salt.
38. The gadolium complex of the compound of claim 37.

39. A complex, or a pharmaceutically acceptable salt of a complex, of a metal atom and a metal chelating ligand having the formula



wherein

- A_1 is $-(CH_2)_m'$ or a single bond;
- $(CH_2)_m$ and $(CH_2)_m'$ may independently be
- 10 substituted with alkyl or hydroxyalkyl;
- R_1 and R_2 are each independently hydrogen,
- $\begin{matrix} S \\ || \\ \text{alkyl}, -NO_2, -NH_2, -NHCNHR_{12}, NCS, -C-NR_3R_4 \text{ and} \\ NR_3COR_9 \text{ where } R_9 \text{ is alkyl or hydroxyalkyl, with the} \\ \text{proviso that at least one of } R_1 \text{ and } R_2 \text{ must be other} \\ 15 \text{ than hydrogen;} \end{matrix}$
- R_3 and R_4 are independently hydrogen, alkyl, arylalkyl, aryl, alkoxy and hydroxyalkyl;
- R_{12} is hydrogen, alkyl or hydroxyalkyl;
- R_{13} is hydrogen, alkyl, arylalkyl, aryl, alkoxy or hydroxyalkyl;
- 20 m and m' are independently 1 to 5; and multimeric forms thereof.
- 40. A complex of claim 39 wherein R_1 and R_2
- 25 are each $\begin{matrix} O \\ || \\ -C-NR_3R_4 \end{matrix}$ wherein each R_3 group is hydroxy-alkyl.

41. A complex of claim 39 wherein R₁ and R₂

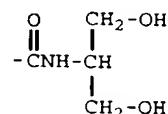
are each $\text{--C}(=\text{O})\text{NR}_3\text{R}_4$ wherein each R₃ group is selected
 from $-\text{CH}_2\text{--CH(OH)--CH}_2$ and $-\text{CH}(\text{CH}_2\text{OH})_2$, and wherein each
 R₄ group is hydrogen.

5 42. A complex of claim 39 wherein R₁ and R₂

are each $\text{--C}(=\text{O})\text{NHCH}_2\text{--CH(OH)--CH}_2$

43. A complex of claim 39 wherein R₁ and R₂
 are each

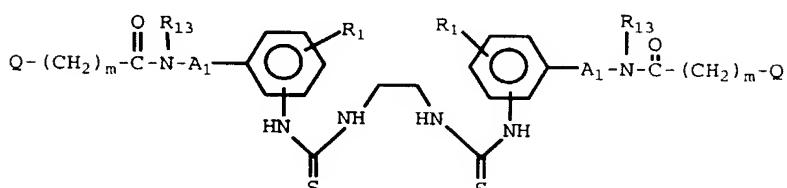
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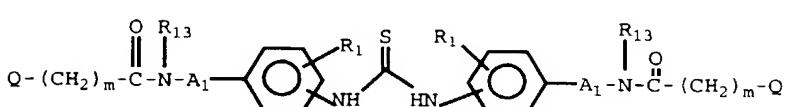
44. A complex of claim 39 wherein said metal
 atom is of atomic number 56-83.

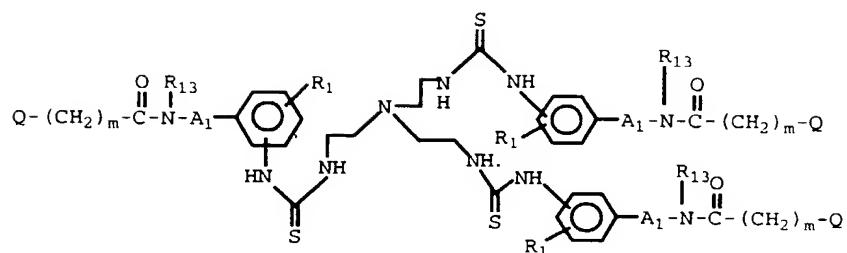
15 45. A complex of claim 39 wherein said metal
 is gadolinium(III).

46. A multimer selected from

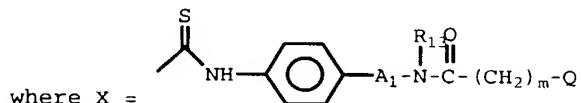
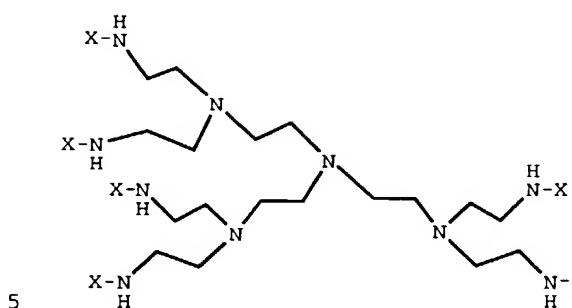


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or



and wherein Q is an aminocarboxylate ligand and the
10 other variables are as defined in claim 3.

47. A compound of claim 6 having the name
10-[2-[[3,5-bis[(2-methylbutyl)amino]carbonyl]-
phenyl]amino]2-oxoethyl]1,4,7,10-tetraazacyclo-
dodecane-1,4,7-triacetic acid.

15 48. The gadolinium complex of the compound of
claim 47.

49. A compound of claim 6 having the name
10,10',10'',10''',10'''',10''''-[[[[[[[[Nitrilo-

tri-2,1-ethanediyl)tris(nitrilo)]hexakis-(2,1-ethane-
diyl)]hexakis(imino)hexakis-(carbonothioyl)]hexakis-
(imino)]hexakis-(4,1-phenylene)]hexakis-(imino))-
hexakis-(2-oxo-2,1-ethanediyl)]hexakis[1,4,7,10-tetra-
5 azacyclododecane-1,4,7-triacetic acid].

50. The gadolinium complex of the compound of
claim 49.